Magnetic precursor effects in Gd based intermetallic compounds

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The behaviour of electrical resistivity (ρ) and magnetoresistance in the vicinity of respective magnetic ordering temperatures in a number of Gd alloys is reported. In some compounds, e.g., $GdNi_2Sn_2$ and $GdPt_2Ge_2$, there is an enhancement of ρ prior to long range magnetic order over a wide temperature range which can be highlighted by the suppression of ρ caused by the application of a magnetic field. However, such features are absent in many other Gd compounds, e.g., $GdCu_2Ge_2$, $GdAg_2Si_2$, $GdAu_2Si_2$, $GdPd_2Ge_2$ and $GdCo_2Si_2$. Attempts to relate such features to magnetic precursor effects in heat capacity are made. On the basis of our studies, we suggest that better understanding of magnetic precursor effects in Gd alloys will be helpful to throw light on some of the current trends in magnetism. Various other interesting findings in the magnetically ordered state in some of these alloys are also brought out.

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I. INTRODUCTION

One of the points of debate in the field of giant magnetoresistance (GMR) is the origin of negative temperature coefficient of resistivity (ρ) above Curie temperature (T_C) and resultant large negative magnetoresistance at T_C [Ref. 1, 2]. Keeping such trends in the field of magnetism in recent years in mind, we have been carefully investigating the magnetoresistance behaviour of some of the Gd alloys in the vicinity of respective magnetic ordering temperatures (T_o), in order to address the question whether such features can arise from some other factor. We have indeed noted an extra contribution to ρ over a wide temperature range above T_o in $GdPt_2Si_2$, $GdPd_2In$, $GdNi_2Si_2$ [Ref. 3], GdNi [Ref. 4], and Gd_2PdSi_3 [Ref. 5], as a result of which the magnetoresistance is negative just above T_o, attaining a large value at T_o, similar to the behavior in manganites. In fact, in one of the Gd compounds, Gd_2PdSi_3, the temperature coefficient of ρ is even negative just above Néel temperature (T_N), with a distinct minimum at a temperature far above T_N. Such observations suggest the need to explore the role of any other factor before long range magnetic order sets in. Similar resistance anomalies have been noted above T_o even in some Tb and Dy alloys.[6] Since critical spin fluctuations may set in as one approaches T_o, the natural tendency is to attribute these features to such spin fluctuations extending to unusually higher temperature range. In our opinion, there exists a more subtle effect, e.g., a magnetism-induced electron localisation (magnetic polaronic effect) and consequent reduction in the mobility of the carriers as one approaches long range magnetic order.[4–6] The efforts on manganites along these lines are actually underway and it appears that a decrease in mobility of the carriers are primarily responsible for negative temperature coefficient of ρ above T_C and large magnetoresistance.[7,8]

The results on the Gd alloys mentioned above are also important to various developments in the field of heavy-fermions and Kondo lattices, as discussed in Refs. 3-5, 9-11. Thus, the investigation of magnetic precursor effects in relatively simple magnetic systems is relevant to current trends in magnetism in general; the Gd systems are simple in the sense that Gd does not exhibit any complications due to double-exchange, crystal-fields, Jahn-Teller and Kondo effects.

We therefore consider it worthwhile to get more experimental information on magnetic precursor effects in Gd systems. In this article, we report the results of electrical resistivity (ρ) measurements in a number of other Gd alloys crystallizing in the same (or closely related) structure, in order to arrive at a overall picture of the magnetic precursor effects in Gd compounds. Among the Gd alloys investigated, interestingly, many do not exhibit such resistance anomalies; in addition, we find that there is no one-to-one correspondence between the (non)observation of excess ρ and a possible enhancement of heat capacity (C) above T_o in these Gd alloys. The compounds[12] under investigation are: $GdCu_2Ge_2$ (T_N= 12 K), $GdAg_2Si_2$ (T_N= 17 K), $GdPd_2Ge_2$ (T_N= 18 K, Ref. 13), $GdCo_2Si_2$ (T_N= 44 K), $GdAu_2Si_2$ (T_N= 12 K),

$GdNi_2Sn_2$ (T_N= 7 K) and $GdPt_2Ge_2$ (T_N= 7 K). While the crystallographic and magnetic behaviour of most of these compounds have been well-known,[12] this article reports magnetic characterization to our knowledge for the first time for $GdNi_2Sn_2$ and $GdPt_2Ge_2$. We have chosen this set of compounds, since all of these compounds are crystallographically related: most of these form in $ThCr_2Si_2$-type tetragonal structure, while $GdNi_2Sn_2$ and $GdPt_2Ge_2$ appear to form in a related structure, viz., $CaBe_2Ge_2$ or its monoclinic modification.[14,15]

II. EXPERIMENTAL

The samples were prepared by arc melting stoichiometric amounts of constituent elements in an arc furnace in an atmosphere of argon and annealed at 800 C for 7 days. The samples were characterized by x-ray diffraction. The electrical resistivity measurements were performed in zero field as well as in the presence of a magnetic field (H) of 50 kOe in the temperature interval 4.2 - 300 K by a conventional four-probe method employing a silver paint for electrical contacts of the leads with the samples; in addition, resistivity was measured as a function of H at selected temperatures; no significance may be attached to the absolute values of ρ due to various uncertainties arising from the brittleness of these samples, voids and the spread of silver paint. We also performed the C measurements by a semiadiabatic heat-pulse method in the temperature interval 2 - 70 K in order to look for certain correlations with the behavior in ρ; respective non-magnetic Y or La compounds have also been measured so as to have an idea on the lattice contribution, though it is not found to be reliable at high temperatures (far above T_o). In order to get further information on the magnetic behavior, the magnetic susceptibility (χ) was also measured in a magnetic field of 2 kOe (2 - 300 K) employing a superconducting quantum interference device and the behavior of isothermal magnetization (M) was also obtained at selected temperatures.

III. RESULTS AND DISCUSSION

The results of ρ measurements in the absence and in the presence of a magnetic field are shown in Fig. 1a below 45 K for $GdPt_2Ge_2$. The C data are shown in Fig. 1b. The χ data in the same temperature interval are shown in Fig. 1c to establish the value of T_N. The magnetoresistance, defined as $\Delta\rho/\rho$= $[\rho(H)-\rho(0)]/\rho(0)$, as a function of H at selected temperatures are shown in Fig. 1d. From the comparison of the data in Figs. a, b and c, it is clear that this compound undergoes long range magnetic ordering at (T_N=) 7 K, presumably of an antiferromagnetic type, considering that the Curie-Weiss temperature (θ_p) obtained from high temperature Curie-Weiss behavior of χ is negative (-8 K) and the isothermal magnetization (M) at 4.5 K does not show indication for saturation (and, in fact, varies linearly with H, Fig. 1c, inset). There is an upturn in ρ below 7 K, instead of a drop, presumably due to the development of magnetic Brillouin-zone boundary gaps;[16] However, with the application of a magnetic field, say 50 kOe, this low temperature upturn in ρ gets depressed; the point to be noted is that there is a significant depression of ρ with the application of H even above 7 K, the magnitude of which decreases with increasing temperature. Thus, there is a significant negative magnetoresistance not only below T_N, but also above it over a wide temperature range. This point can be emphasized more clearly when one measures $\Delta\rho/\rho$ as a function of H at various temperatures (Fig. 1d). There is a quadratic variation with H (up to about 50 kOe) at all temperature mentioned in the plots, attaining a large value at higher fields, and these are characteristics of spin-fluctuation systems. In order to explore whether any such magnetic precursor effects are present in the C data, we show the magnetic contribution (C_m) to C in Fig. 1b after subtracting the lattice contribution (derived from the C data of YPt_2Ge_2) as described in Refs. 9, 17. It appears that this may not be the perfect way of determination of C_m above 30 K as the derived lattice part does not coincide with the measured data for the sample, though the magnetic entropy (obtained by extrapolation of C_m to zero Kelvin) reached its highest value (R ln 8) around 40 K; there may possibly be different degree of crystallographic disorder between Gd and Y alloys, which is responsible for this discrepancy. Clearly the feature is rounded off at the higher temperature side of T_N, resulting in a tail extending to higher temperature range and this feature is free from the error discussed above. The data basically provide evidence for the fact that the full magnetic entropy (R ln8) is attained only in the range 30 - 40 K and it is exactly the same temperature range till which we see an enhancement of

ρ, depressing with the application of H. In short, this compound exhibits magnetic precursor effects both in C and ρ data.

As in the case of $GdPt_2Ge_2$, the results obtained from various measurements for $GdNi_2Sn_2$ are shown in Fig. 2 below 35 K. It is clear from the features in ρ, C and χ that this compound orders magnetically at about 7 K; from the reduced value of peak C_m (lattice contribution derived from the values of YNi_2Sn_2) [Ref. 17] and negative θ_p, we infer that the magnetic structure is of an amplitude-modulated antiferromagnetic type. The main point of emphasis is that there is an excess resistivity till about 15 K, which is highlighted by the depression of ρ with the application of H. Though there are problems similar to $GdPt_2Ge_2$ in deducing precise lattice contribution at higher temperature, we are confident that C_m data (qualitaively) exhibit a tail till about 15 K and the total magnetic entropy is released around the same temperature. The magnetoresistance appears to vary nearly quadratically with H above T_N, say, at 10 and 15 K. Thus, ρ and C data show magnetic precursor effects for this alloy as well.

We now present the results on a series of Gd alloys in which the excess resistance (in the sense described above) is not observable above T_o. These alloys are $GdCo_2Si_2$ (Fig. 3), $GdAu_2Si_2$ (Fig. 4) and $GdPd_2Ge_2$ (Fig. 5). It is clear from the figures 3-5 that the resistivity in the presence and in the absence of H are practically the same (within 0.1%) above their respective ordering temperatures, thereby establishing the absence of an additional contribution to ρ before long range ordering sets in. In order to look for the 'tail' in C_m above T_o, we attempted to obtain respective lattice contributions (employing the C values of YCo_2Si_2, YAu_2Si_2 and YPd_2Ge_2 respectively). We can safely state that the continuous decrease in C_m just above T_o, if exists, does not proceed beyond $1.2T_o$ (See Figs. 3b, 4b and 5b). Thus, it appears that the magnetic precursor effects in C, if present, are negligible, thus tracking the behavior of "excess resistance".

In $GdCu_2Ge_2$ and $GdAg_2Si_2$ as well, clearly there is no excess resistivity above T_N, as the application of H does not suppress the value of ρ (Figs. 6 and 7). However, it contrast to the cases discussed in the previous paragraph, it appears that there is no correlation between C and ρ behavior prior to long range magnetic order. YCu_2Ge_2 and $LaAg_2Si_2$ have been used as references to obtain lattice contributions to C respectively. The finding of interest is that the magnetic contribution to C appears to exhibit a prominent tail (without any doubt in $GdCu_2Ge_2$), at least till 10 K above respective T_N. This behavior is similar to that noted for $GdCu_2Si_2$ earlier.[11,17]

We have also made various other interesting findings:

The peak values of C_m for $GdPt_2Ge_2$ and $GdNi_2Sn_2$ are much smaller than that expected (20.15 J/mol K, Ref. 17) for equal moment (simple antiferro, ferro or helimagnetic) magnetic structures and the fact that the value is reduced by at least a factor of about 1/3 shows that the magnetic structure is modulated. The situation is somewhat similar for $GdPd_2Ge_2$. However, for $GdCo_2Si_2$ and $GdAu_2Si_2$, the peak values of C_m are very close to the expected value for commensurate magnetic structures, thus suggesting that the (antiferromagnetic) magnetic structure is not modulated.

For $GdNi_2Sn_2$ (Fig. 2d), $\Delta\rho/\rho$ as a function of H at 4.5 K exhibits a sharp rise for initial applications of H with a positive peak near 8 kOe. While the positive sign may be consistent with antiferromagnetism, corresponding anomaly in the isothermal magnetization at 4.5 K is not very prominent; the plot of M versus H, however, is not perfectly linear at 4.5 K, showing a weak metamagnetic tendency around 30 kOe (Fig. 2c, inset). It appears that the peak in the magnetoresistance is a result of significant changes in the scattering effects from a weak metamagnetism. Even in the case of $GdAg_2Si_2$, there is a weak feature in the plot of M vs H at 4.5 K around 40 kOe due to possible metamagnetic transition (see Fig. 7d), which is pronounced in the magnetoresistance beyond 20 kOe. In the case of $GdPd_2Ge_2$, at 5 K, $\Delta\rho/\rho$ shows a positive value till 20 kOe, beyond which the value is negative exhibiting a non-monotonic variation with H (Fig. 5d); the plot of M versus H shows only a small deviation from linearity around this field. Thus there are very weak metamagnetic effects which have subtle effects on the scattering processes in the antiferromagnetically ordered state in these compounds. The plot of magnetoresistance versus H and that of isothermal magnetization look similar for $GdCu_2Ge_2$ (Figs. 6d), with a very weak metamagnetic tendency near 35 kOe, as reflected by non-linear plots. These results suggest that the magnetoresistance technique is a powerful tool to probe metamagnetism, even the weak ones, which may not be

clearly detectable by magnetization measurements.

It is to be noted that, interestingly, the value of magnetoresistance is very large at high fields at 5 K (see Fig. 7d) for $GdAg_2Si_2$ (possibly due to granularity?). The heat capacity data in the magnetically ordered state in $GdAg_2Si_2$ as well as in $GdPd_2Ge_2$, reveal the existence of additional shoulders, which may be the result of a combined influence of spin reorientation and Zeeman effects.[17] In particular, for $GdAg_2Si_2$, the magnetic behavior appears to be complex due to the presence of two prominent magnetic transitions, (interestingly) a discontinuous one near 17 K and the other at 11 K (see the features in C and χ in Figs. 7b and 7c). At the 17K-transition in this compound, there is a sudden upward jump in C, and at the same temperature ρ shows a sudden upturn instead of a decrease (Fig. 7), possibly due to the formation of antiferromagnetic energy gaps. It would be interesting to probe whether the transition is first-order in nature. It appears that there is another magnetic transition in $GdCo_2Si_2$ as well around 20 K as seen by an upturn in the susceptibility (Fig. 3c).

IV. CONCLUSIONS

To summarise, on the basis of our investigations on Gd alloys, we divide the Gd compounds into two classes: **Class I**, in which there is an excess contribution to ρ prior to long range magnetic order over a wide temperature range, as a result of which the magnetoresistance is large and negative, e.g., GdNi, $GdNi_2Si_2$, $GdPt_2Si_2$, $GdPt_2Ge_2$. $GdNi_2Sn_2$, $GdPd_2In$, Gd_2PdSi_3. **Class II**, in which such features are absent, e.g., $GdCu_2Si_2$, $GdCu_2Ge_2$, $GdAg_2Si_2$, $GdAu_2Si_2$, $GdCo_2Si_2$, $GdPd_2Ge_2$. (At this juncture, we would like to add that we performed similar studies on compounds like, $GdCu_2$, $GdAg_2$, $GdAu_2$, $GdCoSi_3$ and $GdNiGa_3$ and we do not find any magnetic precursor effects). The present study on isostructural compounds establishes that there is no straightforward relationship between the observation of the excess ρ on the one hand and the crystal structure or the type of transition metal and s-p ions present in the compound on the other. The fact that all the compounds studied in this investigation are of layered type suggests that possible onset of magnetic correlations within a layer before long range magnetic order sets in cannot be offerred as the sole reason for excess resistivity selectively in some cases. If one is tempted to attribute the observation of excess ρ to critical spin fluctuations extending to higher temperature range, as inferred from the tail in C_m above T_o, one does not get a consistent picture, the reason being that, in some of the class II alloys, there is a distinct tail in C_m. It is therefore clear that there must be more physical meaning for the appearance of excess ρ in class I alloys. As proposed in Refs 5 and 6, one may have to invoke the idea of "magnetic disorder induced localisation of electrons" (and consequent reduction in the mobility of the charge carriers) before the onset of long range order, as a consequence of short range magnetic order, detected in the form of a tail in heat capacity. The data presented in this article essentially demand that one should explore various factors determining the presence or the absence of the proposed "magnetic-localisation" effects in the presence of short-range magnetic correlations; possibly, the relative magnitudes of mean free path, localisation length[7] and short range correlation length play a crucial role. It is worthwhile to pursue this question, so as to throw light on several issues in current trends in magnetism.

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FIG. 1. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 45 K as well as the isothermal magnetization (inset) at 5 K for $GdPt_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, as a function of magnetic field (H) at various temperatures is shown in Fig. (d). The lines drawn through the data points serve as guides to the eyes.

FIG. 2. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the derived magnetic contribution (C_m) to C and (c) the magnetic susceptibility for $GdNi_2Sn_2$ below 32 K. The magnetoresistance, $\Delta\rho/\rho$, as a function of magnetic field (H) at various temperatures is shown in Fig. (d). The lines drawn through the data points serve as guides to the eyes. The isothermal magnetization behavior at 4.5 K is plotted in the inset of figure (c) and the low field linear region is shown by a continuous line.

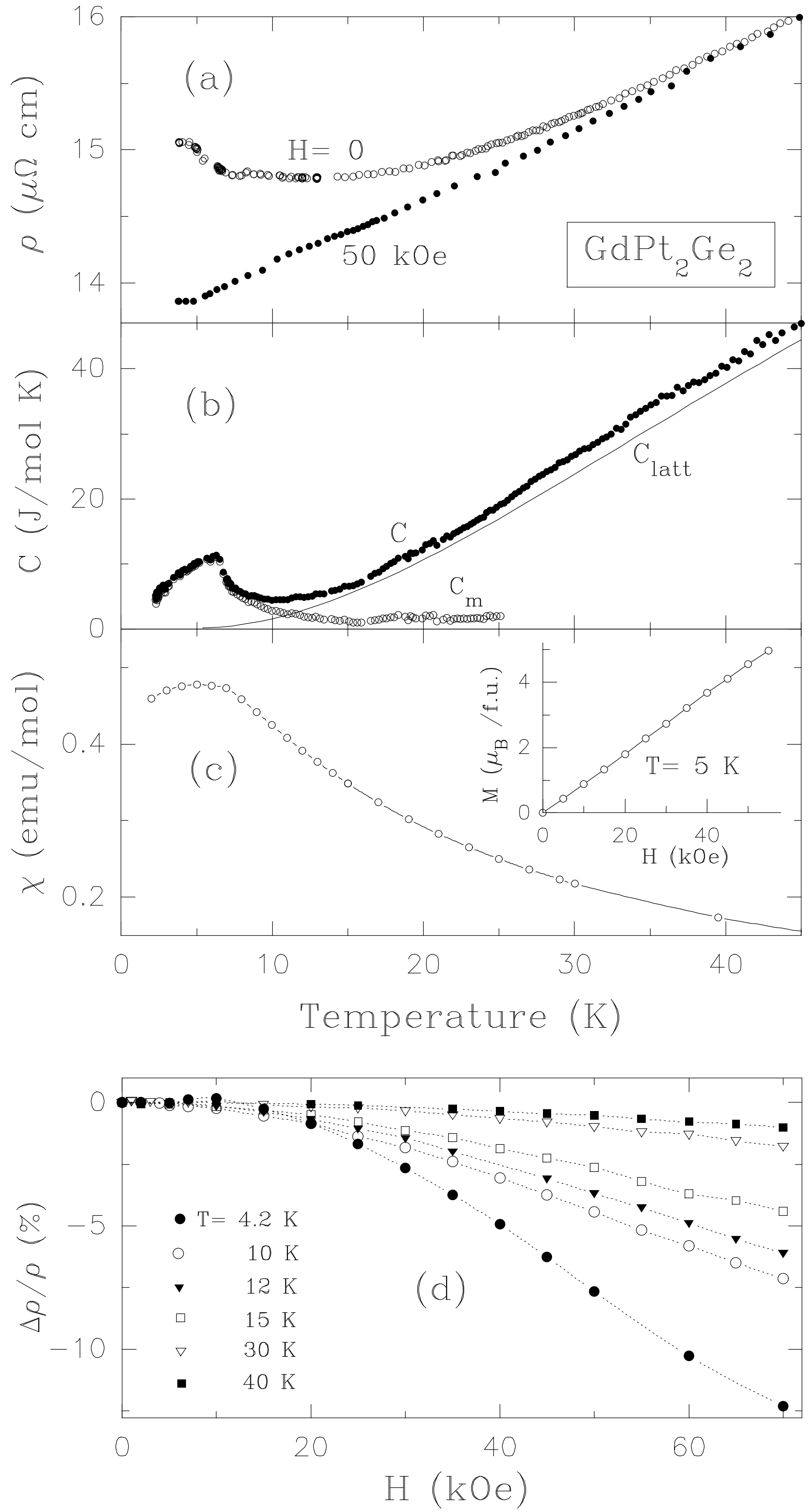
FIG. 3. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 60 K as well as the isothermal magnetization (inset) at 4.5 K for $GdCo_2Si_2$. The lines drawn through the data points serve as guides to the eyes.

FIG. 4. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, and (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C for $GdAu_2Si_2$ below 30 K.

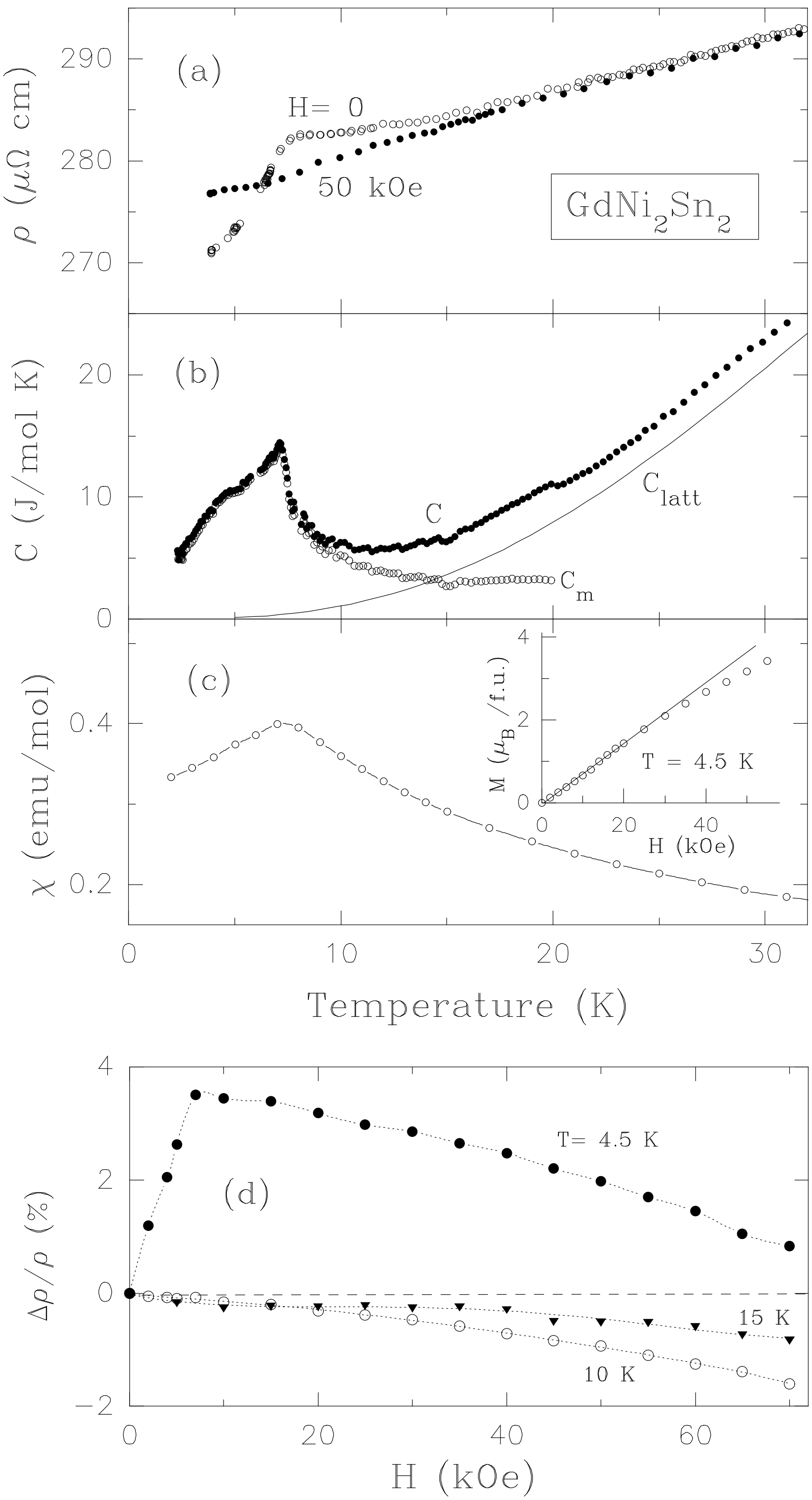
FIG. 5. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 35 K for $GdPd_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetizaton as a function of magnetic field (H) at 5 K are shown in Fig. (d). The lines drawn through the data points serve as guides to the eyes in all the plots except for the M versus H plot, in which case the straight line represents the low field linear region.

FIG. 6. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 30 K for $GdCu_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetization as a function of magnetic field (H) at 4.5 K are plotted in Fig. (d). The lines drawn through the data points serve as guides to the eyes.

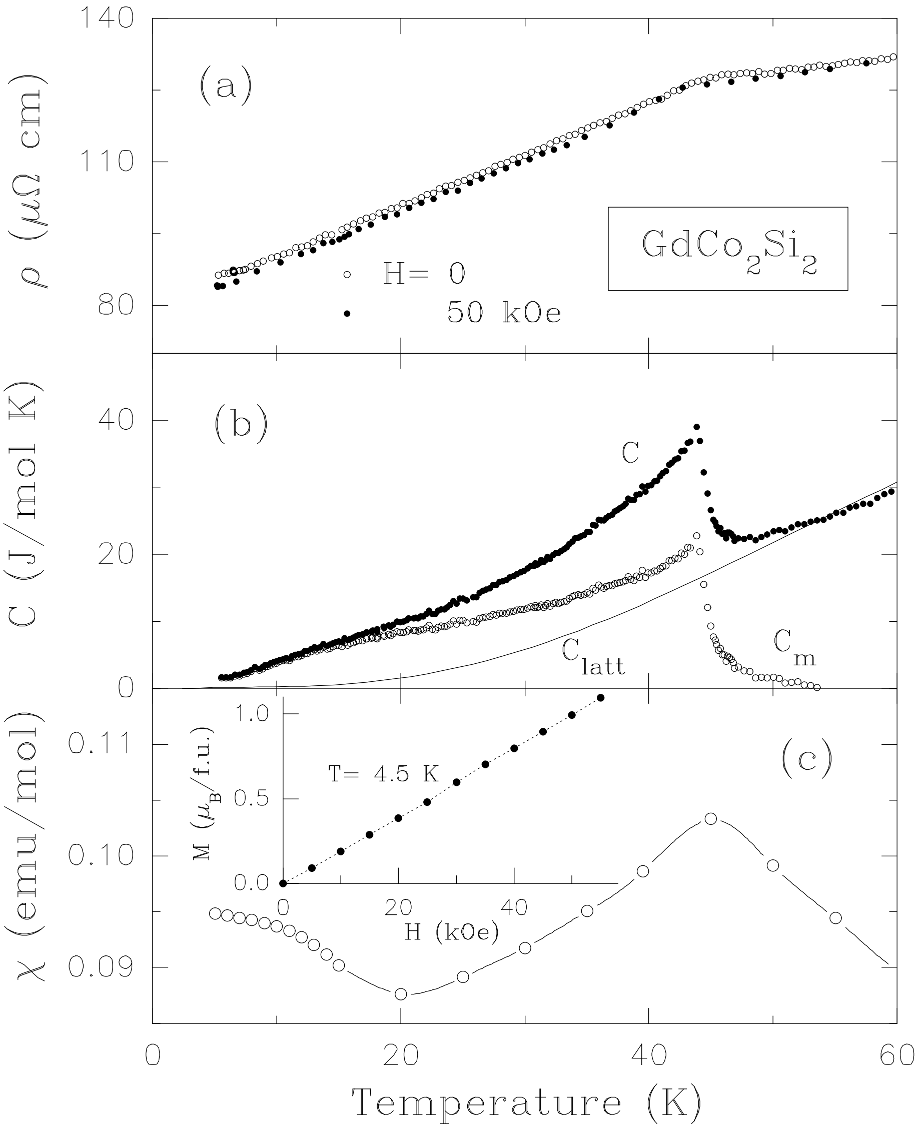
FIG. 7. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to c and (c) the magnetic susceptibility below 30 K for $GdAg_2Si_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetization as a function of magnetic field (H) at 4.5 K are plotted in Fig. (d). The lines drawn through the data points serve as guides to the eyes.



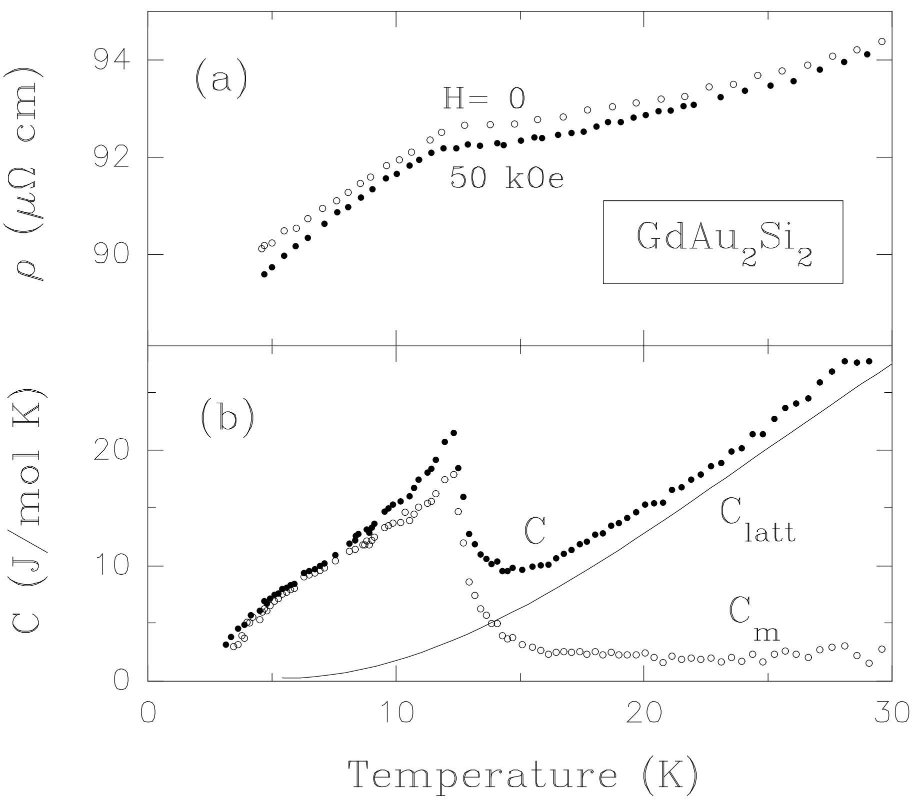
(a)
H= 0
50 kOe
GdPt2Ge2
ρ (μΩ cm)
(b)
C
Clatt
Cm
C (J/mol K)
(c)
χ (emu/mol)
M (μB /f.u.)
T= 5 K
H (kOe)
Temperature (K)
T= 4.2 K
10 K
12 K
15 K
30 K
40 K
(d)
Δρ/ρ (%)
H (kOe)



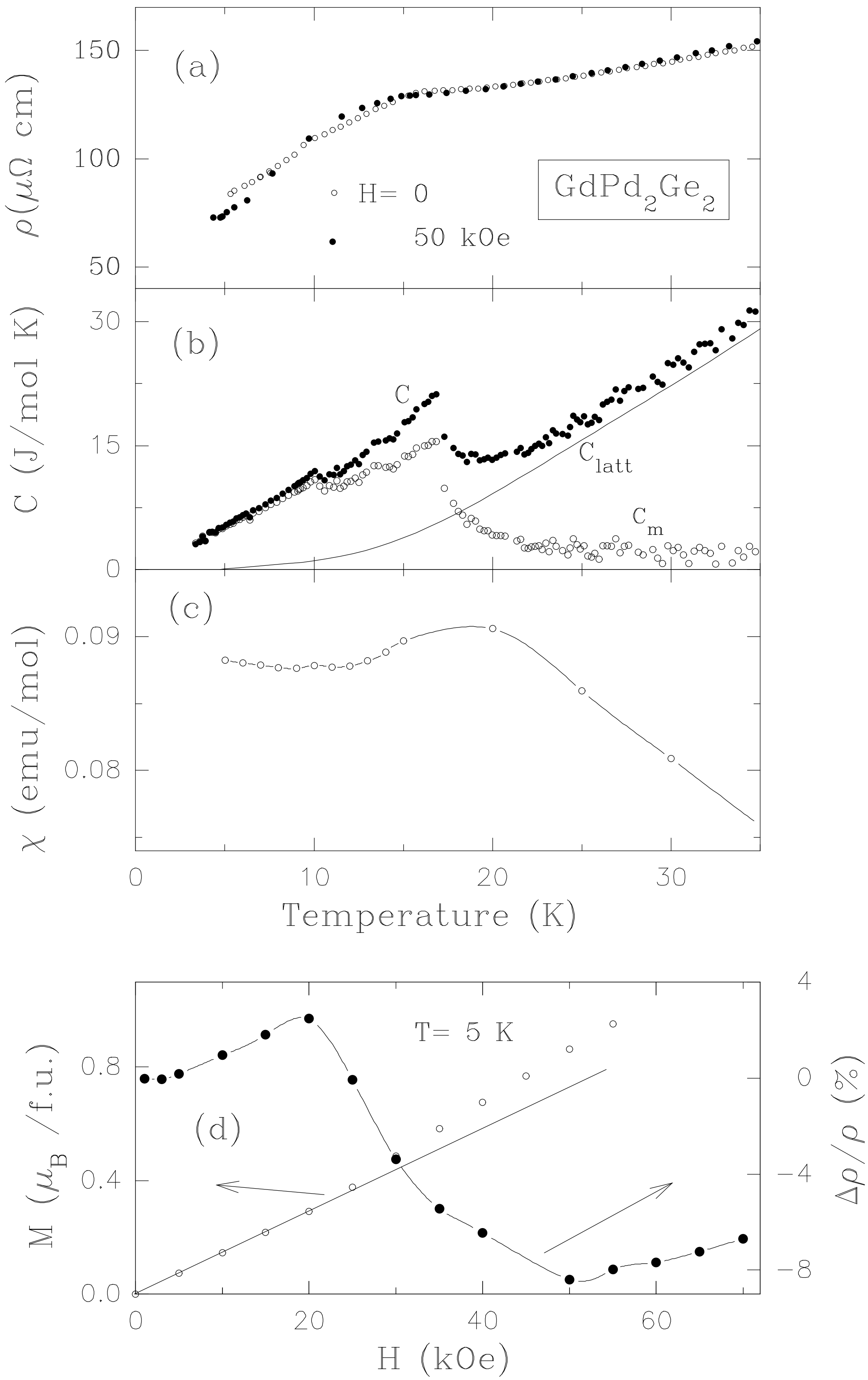
(a)
H= 0
50 kOe
$GdNi_2Sn_2$
ρ ($\mu\Omega$ cm)
(b)
C (J/mol K)
C
C_{latt}
C_m
(c)
χ (emu/mol)
M (μ_B/f.u.)
T = 4.5 K
H (kOe)
Temperature (K)
(d)
$\Delta\rho/\rho$ (%)
T= 4.5 K
15 K
10 K
H (kOe)



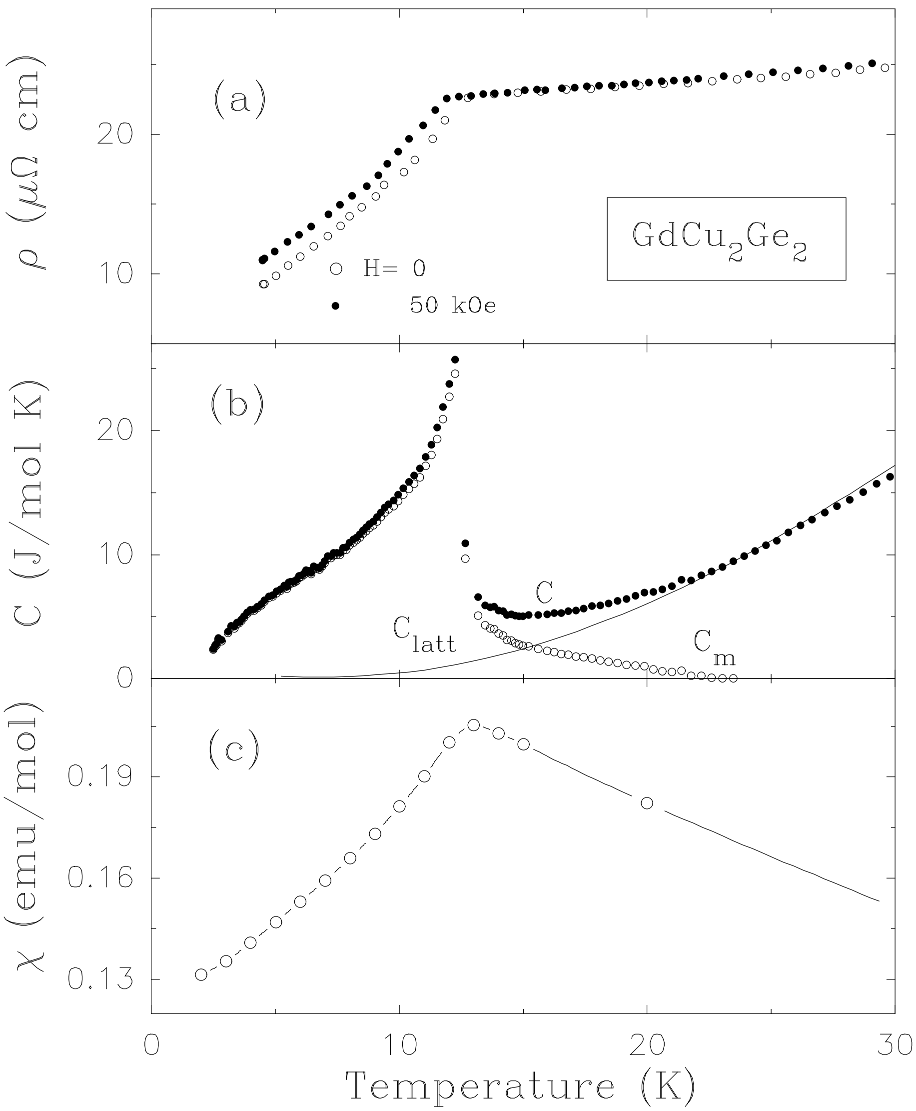
(a)
GdCo2Si2
H= 0
50 kOe
ρ (μΩ cm)
140
110
80
(b)
C
C (J/mol K)
Clatt
Cm
40
20
0
(c)
χ (emu/mol)
0.11
0.10
0.09
T= 4.5 K
M (μB/f.u.)
H (kOe)
Temperature (K)
0
20
40
60



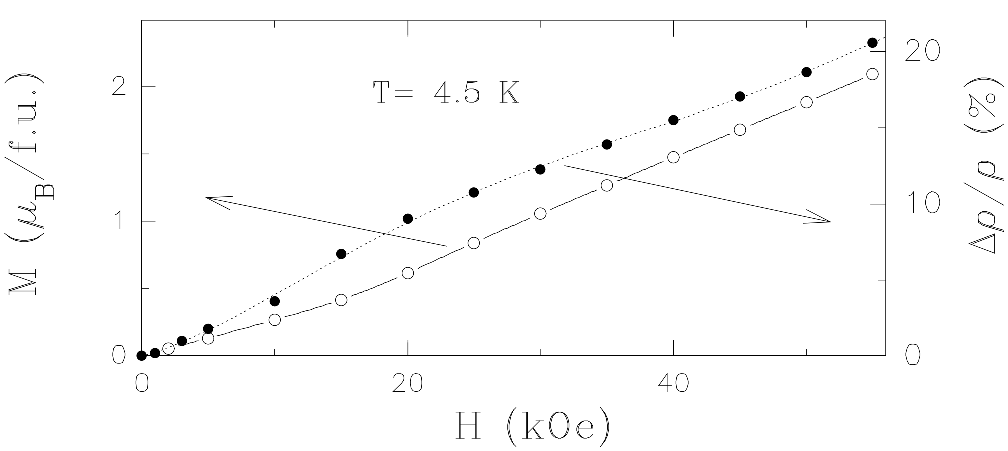
(a)
H= 0
50 kOe
GdAu$_2$Si$_2$
ρ ($\mu\Omega$ cm)
94
92
90
(b)
C
C_{latt}
C_m
C (J/mol K)
20
10
0
0
10
20
30
Temperature (K)



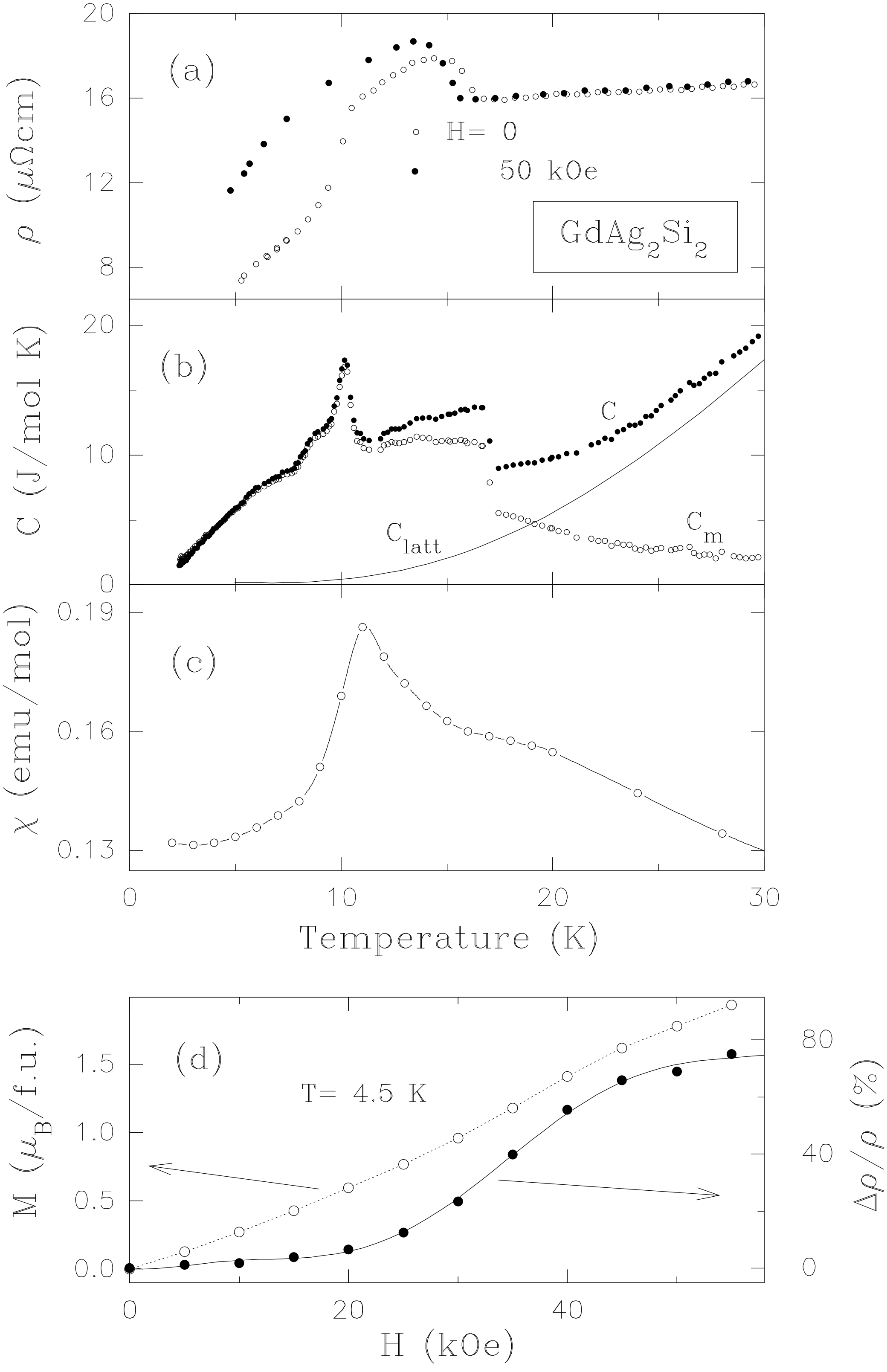
(a)
(b)
(c)
(d)
GdPd$_2$Ge$_2$
H= 0
50 kOe
ρ ($\mu\Omega$ cm)
150
100
50
C (J/mol K)
30
15
0
C
C_{latt}
C_m
χ (emu/mol)
0.09
0.08
0
10
20
30
Temperature (K)
T= 5 K
M (μ_B /f.u.)
0.8
0.4
0.0
$\Delta\rho/\rho$ (%)
4
0
-4
-8
0
20
40
60
H (kOe)



(a)
(b)
(c)
GdCu2Ge2
H= 0
50 kOe
ρ (μΩ cm)
C (J/mol K)
χ (emu/mol)
C
Clatt
Cm
Temperature (K)



T= 4.5 K
M (μB/f.u.)
Δρ/ρ (%)
H (kOe)



(a)
H= 0
50 kOe
GdAg2Si2
ρ (μΩcm)
(b)
C
C latt
C m
C (J/mol K)
(c)
χ (emu/mol)
Temperature (K)
(d)
T= 4.5 K
M (μB/f.u.)
Δρ/ρ (%)
H (kOe)